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E. Merle-Lucotte, D. Heuer, C. Le Brun, M. Allibert, V. Ghetta. The TMSR as Actinide Burner and Thorium Breeder. 2007, 5 p. in2p3-00143680

HAL Id: in2p3-00143680

<https://hal.in2p3.fr/in2p3-00143680>

Submitted on 27 Apr 2007

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The TMSR as Actinide Burner and Thorium Breeder

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INTRODUCTION

Molten Salt Reactors (MSRs) are one of the six systems retained by Generation IV as a candidate for the next generation of nuclear reactors. Molten Salt Reactor is a very attractive concept especially for the Thorium fuel cycle which allows nuclear energy production with a very low production of radiotoxic minor actinides. Studies [1] have thus been done on the Molten Salt Breeder Reactor (MSBR) [2] of Oak-Ridge to re-evaluate this concept. They have shown that the MSBR suffers from major drawbacks concerning for example safety and reprocessing, drawbacks incompatible with any industrial development. On the other hand, the advantages of the Thorium fuel cycle were too attractive not to look further into it.

With these considerations, we have reassessed the whole concept to propose an innovative reactor called Thorium Molten Salt Reactor (TMSR). Many parametric studies of the TMSR have been carried out [3,4,5,6], correlating the core arrangement and composition, the reprocessing performances, and the salt composition. In particular, by changing the moderation ratio of the core the neutron spectrum can be modified and placed anywhere between a very thermalized neutron spectrum and a relatively fast spectrum. Even if the epithermal TMSR configurations have not been completely excluded by our calculations, our studies have shown that the reactor design where there is no graphite moderator inside the core appears to be the most promising in terms of safety coefficients, reprocessing requirements, and breeding and deployment capabilities [7]. Larger fissile matter inventories are necessary in such a reactor configuration compared to the thermalized TMSR configurations, but the resulting deployment limitation could be solved by using transuranic elements as initial fissile load.

This work is based on the coupling of a neutron transport code called MCNP[8] with the materials evolution code REM[1,5]. The former calculates the neutron flux and the reaction rates in all the cells while the latter solves the Bateman equations for the evolution of the materials composition within the cells. These calculations take into account the input parameters (power released, criticality level, chemistry ...), by adjusting the neutron flux or the materials composition of the core on a regular basis. Our calculations rest on a precise

description of the geometry and consider several hundreds of nuclei with their interactions and radioactive decay; they allow a thorough interpretation of the results

1. THE NON-MODERATED THORIUM MOLTEN SALT REACTOR CONCEPT

Core Description

The general TMSR concept is a 2500 MWth (1GWe) reactor operated in the Thorium fuel cycle, using either ^{233}U or Pu as initial fissile matter. As shown in Fig. 1, the core is a single cylinder (1.25m radius and 2.60m height) where the nuclear reactions occur within the flowing salt. One third of the 20 m³ of fuel salt circulates outside of the neutron flux in external circuits flowing through the pumps, the gaseous extraction system which removes the gaseous and non soluble fission products and the heat exchangers before coming back into the core.

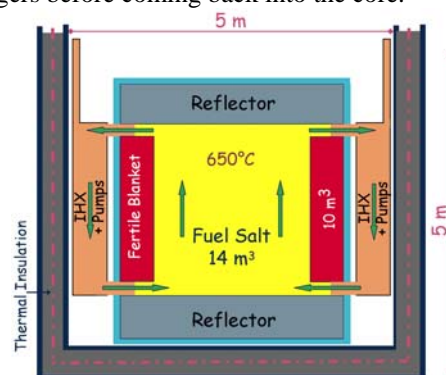


Fig.1. To scale vertical representation of the TMSR, including pumps and heat exchangers (IHx)

The core structures are protected by reflectors which absorb 80% of the neutron flux. To avoid thermalization of the reflected neutrons, the axial reflectors are made of ZrC and not of graphite. The radial reflector consists in graphite channels containing a binary fluoride salt LiF-ThF₄ with 28%- mole ^{232}Th . This reflector, corresponding to a fertile blanket, increases the breeding ratio thanks to ^{233}U extraction every six months.

Salt Reprocessing

The first step consists in the on-line control and adjustment of the salt composition (redox potential measurement, reactivity kept equal to one...).

The salt reprocessing itself is split in two parts. An on-line gaseous extraction system with helium bubbling is dedicated to the removal of gaseous fission products which are strong neutron poisons. As shown during the

MSRE experiment, this bubbling also extract at least some of the noble metals and non soluble fission products. In our simulations, we assume that this helium bubbling is able to extract the gaseous fission products and the noble metals within 30 seconds, a less efficient extraction having little effect on core behavior: up to an extraction time of a few days, the breeding ratio remains nearly stable. The extraction efficiency, which depends on the interaction between the liquid salt, the metallic clusters and the gas bubbles, needs scientific investigations and dedicated measurements to be optimized. Studies are also needed to determine the means to separate the fission products from the gas, to store them and to purify the gas.

For the extraction of the other fission products, mainly lanthanides, a fraction of salt is periodically set aside to be reprocessed off-line. The fissile matter (uranium) can be extracted quickly by fluorination and sent back in the core. The other actinides and lanthanides can be separated via several methods like electrolysis, reduction into metallic solvents, solid precipitation, and any other method studied in the frame of pyrochemistry reprocessing. Finally the actinides are sent back into the reactor core to be burnt, while the lanthanides are stored apart. The performances of the reactor, in terms of breeding and deployment capacities, directly depend on the rate at which this off-line reprocessing is done (see Fig. 2).

Salt Composition

A systematic study has shown that the salt composition (heavy nuclei content) has an influence on neutron energy moderation, actinide solubility, and initial fuel inventory. For heavy nuclei (HN) proportions ranging from 20 to 30 mole%, we choose a binary salt whose melting point is around 570°C, that allows operating at 630°C. For lower proportions of HN, we have either to increase the operating temperature or to add another fluoride to decrease the eutectic point temperature. The salt density ranges from 3.1 to 4.6 according to the HN proportion, with a dilatation coefficient of $10^{-3}/^{\circ}\text{C}$ [9]. The calculations here have been made with a salt containing 80 mole% of LiF completed with BeF_2 but some other possible components (CaF_2 for example) are under study.

2. INITIAL FISSILE CHOICE AND INVENTORIES

TMSR Started with ^{233}U

The ^{233}U initial inventory ranges from 2550 kg for a HN proportion in the salt of 7.5 mole % to 6180 kg for a HN proportion of 27.5 mole % (see Table 1). This corresponds to a variation of the neutron spectrum from an epithermal to a fast spectrum.

Table 1. Initial inventories (kilograms) of Th and fissile matter for ^{233}U -started TMSRs and for transuranic-started TMSRs

| HN proportion | ^{233}U started TMSR | | transuranic-started TMSR | |
|---------------|-------------------------------|------------------|--------------------------|-------------------------------------|
| | Th | ^{233}U | Th | $^{239}\text{Pu} + ^{241}\text{Pu}$ |
| 7.5% | 19760 | 2550 | 13850 | 4887 |
| 10% | 24050 | 3105 | 17560 | 5524 |
| 12.5% | 27790 | 3575 | 20890 | 6022 |
| 15% | 33060 | 4140 | 25380 | 6786 |
| 17.5% | 37230 | 4650 | 29150 | 7297 |
| 20% | 42380 | 5170 | 33640 | 7968 |
| 22.5% | 46100 | 5580 | 37040 | 8378 |
| 25% | 48640 | 5820 | 39320 | 8668 |
| 27.5% | 52190 | 6180 | 42570 | 9037 |

TMSR Started with Transuranic Elements

The initial load of the non-moderated TMSR presented in section 1 can be a mixture of thorium and, for its fissile material, the transuranic elements (Pu, Np, Am and Cm) produced in the water moderated reactors fed at present with natural or slightly enriched uranium. Actually, to be more realistic, these TMSRs are started with the mix of 87.5% of Pu (^{238}Pu 2.7%, ^{239}Pu 45.9%, ^{240}Pu 21.5%, ^{241}Pu 10.7%, and ^{242}Pu 6.7%), 6.3% of Np, 5.3% of Am and 0.9% of Cm, corresponding to the transuranic elements of an UOX fuel after one use in a standard PWR and five years of storage [10].

For a typical TMSR configuration with 17.5 mole% of HN in the salt, an amount of 7300 kg of fissile elements is needed initially, corresponding to 4.5 mole % of Plutonium (See Table 1). This is equivalent to the fissile matter contained in a fast neutron reactor (FNR), a single load being needed in our case while two such loads are necessary for a FNR.

3. CHARACTERISTICS OF THE TMSR AS THORIUM BREEDER AND ACTINIDE BURNER

Influence of the Off-line Reprocessing

For each proportion of heavy nuclei in the salt, ranging from 6 mole % to 27.5 mole %, we have evaluated by simulation the breeding ratio of each reactor configuration as a function of the amount of heavy nuclei reprocessed per day. The result of this study is displayed in Fig. 2, allowing the visualization of many parameters of interest of the non-moderated TMSR: reprocessing design, reactor deployment and breeding capacities. Our calculations have been done for several realistic reprocessing rates, ranging from 50 to 200 kg of HN reprocessed per day.

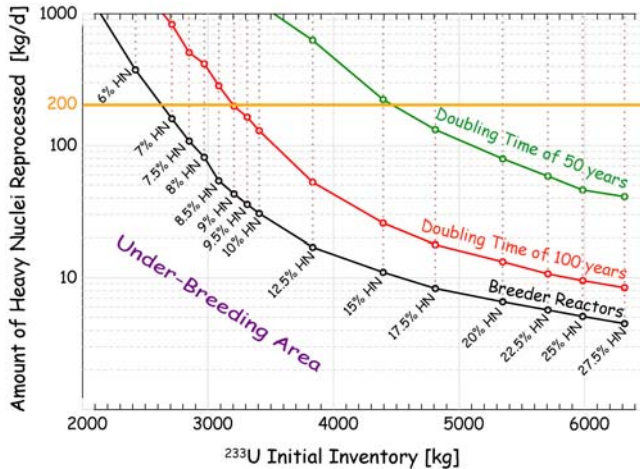


Fig.2. Amount of heavy nuclei reprocessed per day versus initial fissile (^{233}U) inventory, for different heavy nuclei proportions in the fuel

Deterministic Safety

The total feedback coefficient at equilibrium is displayed in Fig. 3, together with its components, the contributions of the salt heating and salt density, as a function of the HN proportion in the salt. All these safety coefficients are significantly negative for all HN proportions, including the density coefficient which can be viewed as a void coefficient. The total feedback coefficient ranging from -10 pcm/K to -5 pcm/K and thus ensuring a very good level of deterministic safety[11] in all these ^{233}U -started TMSR configurations, safety is not a discriminating factor in choosing the optimal salt composition.

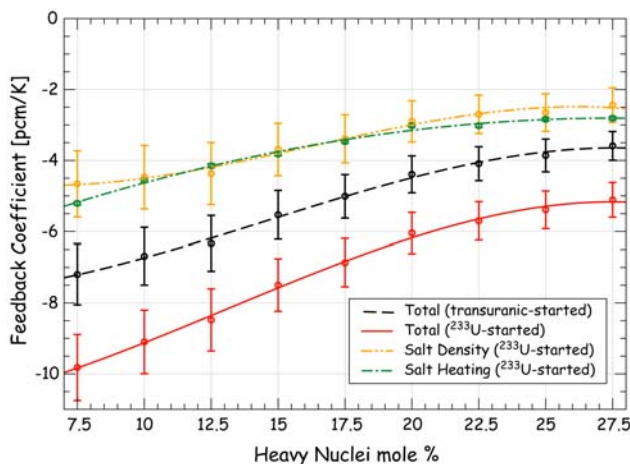


Fig.3. Feedback Coefficients of ^{233}U -started TMSRs (at equilibrium) and of transuranic-started TMSRs (after one year of operation) as a function of the HN proportion

Concerning TMSRs started with transuranic elements, the total feedback coefficients after one year of operation are also displayed in Fig. 3 as a function of the HN proportion. They correspond to the initial safety

behavior of the reactor, since the inventories after one year of operation are quite similar to the initial inventories, but we also take into account the effects of the fission products which are not present in the initial load of the core. As shown in Fig. 3, the safety coefficients are initially slightly smaller in the case of the transuranic-started TMSRs but they remain largely negative, being equal at equilibrium to the safety coefficients of the ^{233}U -started TMSRs.

Heavy Nuclei Inventories

Fig. 4 illustrates the evolution of a typical fuel salt composition all along the operation of this reactor, for the ^{233}U -started (solid lines) and for the transuranic-started (dashed lines) TMSR. At equilibrium, the $^{232}\text{U}/^{233}\text{U}$ ratio in the fuel salt is around 0.1 to 0.3%, corresponding to 2.5 to 20 kg for HN proportions ranging from 7 to 27.5 mole %. This ^{232}U is a major asset against proliferation in the case of the Th- ^{233}U cycle, while presenting no major disadvantage during reprocessing thanks to the liquid fuel.

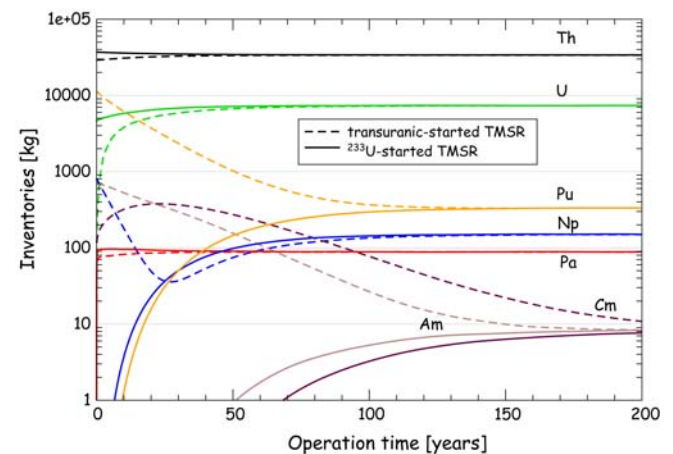


Fig.4. Heavy nuclei inventory for the ^{233}U -started TMSR (solid lines) and for the transuranic-started TMSR (dashed lines) with 17.5 mole% of heavy nuclei in the salt

In terms of transuranic inventory, as shown in Fig. 4, the TMSRs started with transuranic elements become equivalent to TMSRs directly started and operated with ^{233}U after about forty years for a fuel salt with 17.5% of heavy nuclei, where more than 85% of the initial TRU inventories are burned. More generally, the assets of the Thorium fuel cycle are finally recovered for these transuranic-started TMSRs after 25 to 50 years for HN proportions ranging from 7 to 27.5 mole %.

Deployment Capacities

The operating time necessary to produce one initial fissile (^{233}U) inventory is called the reactor doubling time.

Transuranic-started TMSRs allow the extraction of significantly larger amounts of ^{233}U during their first 20 years of operation, thanks to the burning of TRUs which

saves a part of the ^{233}U produced in the core. These higher deployment capacities allowed by the use of TRUs in the transuranic-started TMSR are visible on the reactor doubling times, displayed in Fig. 5 (dashed line), where the configurations with HN proportions larger than 15% have the lower reactor doubling times, around 30-35 years, to be compared with the doubling times greater than 45-50 years reachable with the ^{233}U -started TMSRs.

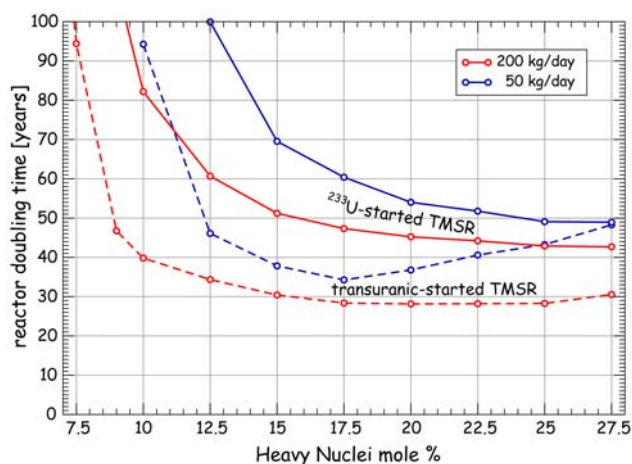


Fig.5. Reactor doubling time of ^{233}U -started (solid lines) and transuranic-started TMSRs (dashed lines) for the different HN proportion configurations and for two off-line reprocessing schemes: 200 kg (red lines) and 50 kg (blue lines) of HN reprocessed per day.

The configuration with 17.5% of heavy nuclei in the salt appears particularly optimal in terms of deployment capacities, especially with the faster reprocessing scheme and for the transuranic-started TMSRs.

CONCLUSION

We have presented here a very promising, simple and feasible concept of Molten Salt Reactor with no moderator in the core, called non-moderated Thorium Molten Salt Reactor (TMSR).

TMSRs benefit from several specific advantages, mainly due to the liquid fuel and to the Thorium cycle. The liquid fuel does not have to be kept under high pressure even at high temperature; it is very stable vis-à-vis irradiation and it avoids the fuel refabrication problem in the presence of actinides. Moreover, using a liquid fuel allows fuel control to be carried out regularly. The amounts of fissile and fertile matter can be adjusted without unloading the core, doing away with the need for any initial reactivity reserve.

The non-moderated TMSR configurations with high HN proportions present particularly interesting characteristics concerning their safety performances and their ability to be first loaded with transuranic elements produced by the

current water reactors. Finally, their rather large initial fissile inventory does not inhibit their capability for a fast deployment thanks to their very good ^{233}U breeding and they are non proliferating because of the presence of ^{232}U .

Under these conditions the TMSR appears to be a very appealing concept; building the reactor is not very complicated, and our calculations do not indicate a major reprocessing constraint, allowing batch mode reprocessing in the vicinity of the reactor. The main studies needed to demonstrate the scientific feasibility of the concept deal with the on-line control of the salt composition and of its chemical and physical properties. Such studies are in progress in the frame of the French concerted research program 'Molten Salt Reactor' (PCR-RSF). Finally we want to point out the hardiness and the flexibility of this TMSR concept, allowing it to be adjustable without losing its advantages in the event of any technologic issue.

All these properties put the TMSR in a very favourable position to fulfil the conditions defined by the GEN IV International Forum and to produce the large amount of nuclear energy that the world will need in the near future.

ACKNOWLEDGMENTS

We are thankful to Elisabeth Huffer for her help during the translation of this paper.

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